

#### ADDITIONAL FEE

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#### REMARKS

The applicants would like to thank Examiner Lovering for his suggestions to overcome the 35 U.S.C 112, second paragraph rejections. These recommendations have been incorporated into claims 2 and 3. It is believed that no new matter has been added. Claims 1-3 are pending.

The remaining issue at hand is the rejection of claims 1-3 over Allard et al. (U.S. Patent 5,616,331). The applicants respectfully request reconsideration of the previous arguments in light of the additional comments below.

One of the key points relied upon by the examiner in the Allard et al. reference is that Allard et al. teaches a *microemulsion*. However, it does not appear that Allard et al. in fact actually formed a microemulsion (they refer to their invention as an "ultrafine" emulsions).

The applicants define the size range of their microemulsion (see page 2, lines 23-25) to be  $10^{-2}$  to  $10^{-1}$   $\mu\text{m}$  (10 to 100 nm or 100 to 1000 Angstroms) which is consistent with the generally accepted meaning of the term (see e.g. page 143 from *Emulsions and Emulsion Technology (Part III)*, Marcel Dekker, Inc., (1984) and Microemulsion: A Definition (from the Surfactants Virtual Library - <http://surfactants.net/microemulsion.htm>, (2002)).

Another property of microemulsions are that they are clear solutions ("transparent or translucent"). Whereas the applicants invention is transparent/translucent due to the fact that their compositions are microemulsions, the emulsions of Allard et al. appear to be transparent/translucent due to the addition of nanopigments which is stated by Allard et al. to be an essential feature of their invention, i.e. any transparency disclosed by Allard et al. is due to the

content of the ingredients in their emulsion, *not because their composition is a microemulsion.*

Allard et al. (col. 5, lines 58-63) describes the size of the particles in their invention as follows:

"According to an essential feature of the present invention, the average size of the liquid particles (or globules) of the fatty phase dispersed within the aqueous dispersing phase must satisfy very particular limits, namely ranging from 100 nm to 1,000 nm. Preferably this average size ranges from 100 nm to 500 nm."

It is unclear that the lower value (i.e. 100 nm) of Allard et al.'s range is intended to be included as col. 6, lines 13-17 states:

"The emulsifying systems which must thus be used are those which actually permit stable ultrafine emulsions resulting from phase inversion ( $100 \text{ nm} < \phi_{\text{globules}} < 1000 \text{ nm}$ ) to be obtained, and in which the nanopigments are dispersed finely and homogeneously."

Further indication, that the emulsions of Allard et al. have particle/globule sizes greater than 100 nm and are not a microemulsion, is the fact that Allard et al.'s examples only state that the emulsion they formed had a globule size of  $< 1 \text{ } \mu\text{m}$  (1000 nm) and that there was the absence of agglomerates larger than  $3 \text{ } \mu\text{m}$  (3000 nm) in size (which obviously suggests that agglomerates of less than 3000 nm were present which would be inclusive of particle/globule sizes which are characteristic of an emulsion) - see col. 8 Table I, example F1. Allard et al. refers to their invention as "ultrafine" *emulsions* and pointedly avoids ever calling their preparations to be "microemulsions" even though this term was well known in the art at the time of filing of Allard et al.'s invention (foreign priority date is 9 February 1994 - nearly ten years after the publication of *Emulsions and Emulsion Technology (Part III)*).

Given that the state of the art is such that microemulsions and emulsions are compositions with distinctly different characteristics, one of ordinary skill in the art would not be directed or motivated by the teachings of Allard et al. to obtain the applicants claimed microemulsions. At best, it appears that Allard et al. teaches how to make emulsions which mimic some of the properties of microemulsions while still remaining an emulsion composition.

***Closing***

Applicants believe that the foregoing constitutes a bona fide response to all outstanding objections and rejections.

Applicants also believe that this application is in condition for allowance. However, should any issue(s) of a minor nature remain, the Examiner is respectfully requested to telephone the undersigned at telephone number (212) 808-0700 so that the issue(s) might be promptly resolved.

Respectfully submitted,

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I hereby certify that the foregoing Amendment Under 37 CFR § 1.116 (After final response) is being deposited with the United States Postal Service as first class mail in an envelope addressed to: Hon. Commissioner of Patents, Washington, D.C. 20231, on the date indicated below:

Date: **26 February 2002**

By Howard C. Lee  
Howard C. Lee

**Microemulsion: A Definition**

A microemulsion is a thermodynamically stable dispersion of one liquid phase into another, stabilized by an interfacial film of surfactant. This dispersion may be either oil-in-water or water-in-oil. Microemulsions are typically clear solutions, as the droplet diameter is approximately 100 nanometers or less. The interfacial tension between the two phases is extremely low.

Emulsions (or macroemulsions) are in contrast unstable, the suspended droplets will eventually agglomerate and the dispersed phase will phase separate. Emulsion droplet sizes are much larger, typically one micron or more, resulting in a cloudy or milky dispersion. The nature of an emulsion may depend on the order of mixing of the ingredients and the amount of energy put into the mixing process. The final microemulsion state will not depend on order of mixing, and energy input only determines the time it will take to reach the equilibrium state.

Microemulsions are two phase systems, in contrast to micellar solutions, which may be considered one phase.

For books on microemulsions and their applications, click [here](#).

For more information on surfactants, see the [Surfactants Virtual Library](#).



# EMULSIONS AND EMULSION TECHNOLOGY

(IN THREE PARTS)

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## PART III

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Library of Congress Cataloging in Publication Data  
(Revised for volume 3)

Main entry under title:

Emulsions and emulsion technology.

(Surfactant science series, v. 6)

Includes bibliographical references and indexes.

1. Emulsions. I. Lissant, Kenneth J.

TP156.E6L57      660.2'84292      73-82192

ISBN 0-8247-7083-8

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MARCEL DEKKER, INC.

270 Madison Avenue, New York, New York 10016

Current printing (last digit):

10 9 8 7 6 5 4 3 2 1

PRINTED IN THE UNITED STATES OF AMERICA

standpoint of stability is discussed by Becher [7]. It has long been known that to prepare a stable emulsion, a third component of a particular type has to be added. These components are inorganic electrolytes, surface-active compounds of the soap, and macromolecular emulsifying agents (gum, starch, protein, etc.) [8]. Investigations on the effect of surface-chemical factors on stability, application of the theory of stability of lyophobic colloids to emulsions, and stability against coalescence related to mechanical properties of interfacial layers have been reviewed by Kitchener and Mussellwhite [8].

Van den Tempel [9], Greenwald [10], and Sonntag et al. [11] studied the stability of oil-in-water emulsions on the basis of the Debye-Huckel model of the electrical double layer. In water-in-oil emulsions, however, the effect of solvation is more efficient than the electrostatic charge in stabilizing the system [12]. The low dielectric constant will make the electrical potential more diffused, and there will be no significant electrostatic potential barrier to prevent the water droplets from coming close to each other. According to Cockbain and McRoberts [13], the stability is caused by the wetting resistance of segments of adsorbed film. This will depend on temperature, particle size, viscosity of the external phase, and the nature of the adsorbed film which has to be displaced from the surface to cause coalescence. Using multicomponent systems, Schulman and Cockbain [14] found that to obtain a stable water-in-oil emulsion, the emulsifying agent must be brought to the interface and form a rigid film. One surfactant type will tend to fit between the positions of the other different surfactant, giving the effect of packing the surface and thereby improving the stability of the emulsion droplets and thus preventing coalescence.

### III. MICROEMULSIONS—THEORETICAL CONSIDERATIONS

We shall now examine the historical developments in the area of emulsions leading to the birth of microemulsions. The current views on the formation, structure, and stability of emulsions related to tertiary oil recovery will be discussed in this section.

#### A. Historical Developments

To earlier workers such as Clowes, Bancroft, Donnan, Harkins, Hildebrand, or Pickering and even to such recent investigators as Schulman, Cockbain, and Clayton [7,15], emulsions were systems which rapidly separated into the original two phases and appeared milky white. Exceptions were the transparent systems (oil-in-water or water-in-oil microdroplets) which were later called *solubilized systems*. As time passed, more extensive studies were made on these systems. From the studies on stability, phase inversion, and interfacial tension measurements, Schulman and co-workers [14,16] concluded that the

reactions occurring at the oil/water interface were closely analogous to the corresponding reactions at an air/water interface. Hoar and Schulman [17] later described the transparent oil-water dispersions as "oleopathic hydromicelles," which are now referred to as *microemulsions*. The spherical oleophilic hydromicelle and the hydrophobic oleomicelle [18] soon became the accepted models for the water-in-oil and oil-in-water emulsion systems. These models suggested that an increase in the total interfacial area involves the transition from macroemulsion to microemulsion. Studies on the effect of chain length and structure of alcohol by Schulman and McRoberts [19] using electrical conductivity measurements suggested that the alcohol molecules must associate with the soap molecules at the interface and change the wettability of the interface by the oil and water according to the hydrophilic-lipophilic balance of the interfacial film. The next 17 years from 1943 to 1960 saw a period of development for the transparent oil and water systems. The structure and properties of microemulsions were investigated by Schulman and co-workers, who used various physical techniques such as low-angle x-ray measurements [20], light scattering [21], ultracentrifugation [22], and electron microscopy [23,24]. Schulman et al. [23] used the term *microemulsions* to describe such transparent oil/water dispersions. They concluded that microemulsions are isotropic, clear or translucent, thermodynamically stable dispersions of oil, water, and emulsifiers with the droplet size ranging from 100 to 1000 Å in diameter. Since then, extensive studies were carried out on the various aspects of microemulsions by several investigators. A more detailed picture of the earlier work can be obtained from the reviews by Prince [25,26], Friberg [27], and Shinoda and Friberg [28].

## B. Recent Developments

During the last two decades, microemulsions have been studied extensively, both theoretically and experimentally, by various investigators [29-46]. Alternate terminologies have also been proposed by different workers for microemulsions [30,34,36,39,42,47]. Shah and co-workers [32,33,37,41,48] showed that the microemulsion undergoes a series of structural changes as the amount of water is increased. Using various physical techniques such as birefringence, electrical resistance, high-resolution NMR, spin-spin relaxation time, and viscosity measurements, they have shown the existence of structurally dissimilar microemulsions [49]. Studies on the structural aspects of systems containing oil, surfactant, and water using dielectric and ESR spin-labeling techniques have been reported by Clausse et al. [50], Eicke and Shepherd [38], Menger et al. [51], and Kitahara et al. [52].

Apart from Schulman's four-component microemulsion systems, three-component systems containing water, oil, and surfactant or water, oil, and cosurfactant (alcohol) are also known. These systems are